

Form birefringence and negative index change created by femtosecond direct writing in transparent materials

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Although femtosecond lasers have proved to be of great utility for micromachining within bulk transparent materials, little is known about the fundamental physics that drive the process. Depending on the laser intensity delivered to the sample, any of three types of feature can be written into the glass. We observed that in the intermediate regime there is a correlation among the negative sign of the effective index change, the presence of anisotropic reflection, and birefringence. We propose a model that can explain all three principal characteristics. Results show that the local index change can be as high as 10^{-1} . © 2004 Optical Society of America

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When an intense femtosecond laser pulse is focused into transparent material, high-order nonlinear absorption allows the energy to be deposited predominantly within the focal volume, producing a local permanent refractive-index modification. Although the process of energy absorption is now well understood,¹ little is known about the actual formation of the directly written structures.² Depending on the level of laser intensity, one can induce any of three qualitatively different types of modification or damage^{3,4}: low intensity induces a smooth positive refractive-index change relative to the unprocessed material (type 1); intermediate intensity results in birefringent regions (type 2); and at high intensity the damage consists of voids embedded into the glass (type 3). The transition threshold from one kind of structure to another is not specified here, as different groups of investigators did not process the same glass and because it is well known that the birefringence in femtosecond directly written samples is onset at a certain intensity threshold that depends on the processed material and on the laser's pulse duration and wavelength.^{4,5}

Although the full range of characteristics that one can create with femtosecond direct writing remains largely unexplored, important applications have already been demonstrated. Type 1 modification has been used to achieve waveguides and couplers,^{2,6} whereas type 3 voidlike defects have been exploited for data storage and photonic crystals.⁷ The intermediate type 2 regime has received little attention until now but indeed seems to have intriguing properties. Type 2 structures in Ge-doped silica show an anisotropic light scattering that is dependent on the plane of polarization of the light.⁸ Anisotropic scattering has been interpreted in terms of photoelectrons moving along the direction of the light's polarization, inducing index inhomogeneities. More recently, uniaxial birefringence imprinted in structures written within fused-silica plates was observed.⁴ We then discovered anisotropic reflection, which we explained in terms of the formation of a self-organized nanograting within the focal volume,⁵ and recently the

existence of the self-induced nanograting was proved by direct imaging.⁹ We point out in this Letter that this nanograting is also associated with a negative effective index change, and we provide a consistent explanation for all the effects named above, using a model of form birefringence.

An amplified, mode-locked Ti:sapphire laser operating at a wavelength tunable from 800 to 850 nm, with a 200-fs pulse duration and a 250-kHz repetition rate, was utilized to effect all the direct-write processes. The laser light was focused via either a $10\times$ (N.A., 0.21) or a $50\times$ (N.A., 0.55) objective to a focal spot of ~ 4 or $\sim 1.5\ \mu\text{m}$, respectively, into the sample, which was mounted upon a computer-controlled linear motor translation stage. Using this setup with the laser tuned at 835 nm and focused by the $10\times$ objective, we directly wrote a matrix of 4×4 quasi-uniform regions of $100\ \mu\text{m}\times 100\ \mu\text{m}$ each (sample 1) within the bulk of silica glass (HeraSil 1). We constructed each processed zone by writing 100 adjacent lines spaced $1\ \mu\text{m}$ apart. To be able to find the transition between types 1 and 2 of damage, we wrote the micromachined zones with different laser dosages. Each row of the matrix of quasi-uniform regions was processed with constant average power ($P1 = 160\ \text{mW}$, $P2 = 100\ \text{mW}$, $P3 = 80\ \text{mW}$, $P4 = 60\ \text{mW}$); each column, with the same number of pulses ($N1 = 5\times 10^4$, $N2 = 2.5\times 10^4$, $N3 = 1.7\times 10^4$, $N4 = 1\times 10^4$). Figure 1 shows an image of the first two rows of the sample acquired between cross polarizers. Only the first row ($P1$) of the matrix, written at the highest intensity, shows the birefringence that we describe as type 2; the rest are of type 1.

To facilitate the measurement of the refractive-index change of each zone with respect to the unprocessed bulk, we used an interferometric phase-stepping technique.¹⁰ Figure 2 shows a schematic of the interferometer with which we measured the laser-induced phase shift. The sample was positioned in the fixed arm of the interferometer, and we varied the reference arm by moving mirror M2 by using a computer-controlled piezoelectric actuator. A polarized He-Ne laser beam was split by optical flat OF1

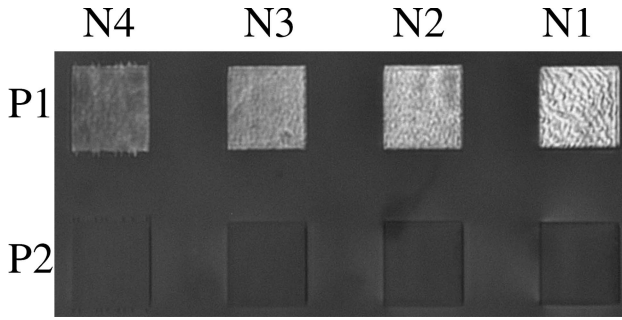


Fig. 1. Picture of part of sample 1 taken between cross polarizers.

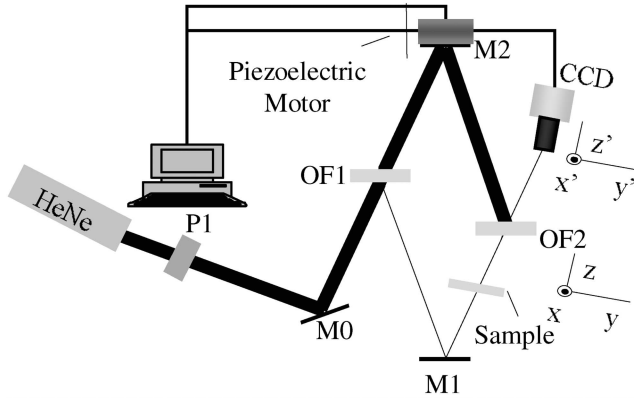


Fig. 2. Schematic of the interferometer used to measure the phase shift of the directly written sample with respect to the untouched bulk: M0–M2, mirrors; P1, polarizer; other abbreviations defined in text.

at the input of the interferometer, recombined at OF2, and imaged by a charged-coupled device (CCD), which transferred the data to a computer. Intensity I at the CCD camera was therefore given as

$$I(x', y') = I_0(x, y) \{1 + V \cos[\phi(x, y) + \phi_0(t)]\}, \quad (1)$$

where (x, y) was the plane where the sample sat and (x', y') was its conjugate plane at the CCD camera, I_0 was the bias in the interference fringe, and V was the visibility of the fringes. ϕ_0 was the variable phase, and ϕ was the additional unknown phase added by the sample. ϕ_0 was stepped through approximately 2π with a CCD image captured at each step. The intensity at each pixel of the CCD evolved according to Eq. (1), which we then fitted to extract relative phase ϕ , resulting in the phase image shown in Fig. 3. This setup permitted a resolution of $\sim 5 \mu\text{m}$ and a level of noise of ± 0.1 rad.

Figure 3 shows the phase images of the directly written sample for light traveling in the interferometer polarized parallel to the writing light [xx ; Fig. 3(a)] and orthogonal to it [xy ; Fig. 3(b)]. Table 1 lists the measured relative phase shift versus polarization for each region of the matrix, with strong birefringence apparent in P1. To obtain more data we also analyzed the birefringent directly written Fresnel zone plates A and B previously reported.¹¹ In this case we measured $\Delta\phi_{xx} = (-1.5, -1.3)$ rad, respectively,

and $\Delta\phi_{xy} \sim -0.2$ rad for both plates. The striking result is that the xx -polarized light traveling into the samples sees a negative phase shift with respect to the light traveling into untouched silica. The xy -polarized light sees a small negative index change in the Fresnel zone plates and a positive index change in sample 1.

We found the association of the negative index change with the birefringence to be consistent with our previous explanation of the formation of a nanograting within the focal volume.⁵ This structure has been interpreted as a self-organized, periodic refractive-index modulation produced by a pattern of interference between the incident laser radiation and an electron acoustic wave generated within the sample.⁹ By applying a theoretical model of form birefringence to the directly written structures we can justify the negative index change as well as the possibility of a polarization-dependent sign of the effective index change.

The inset of Fig. 4 shows an idealized schematic of the self-organized nanograting induced in the glass, where $T = t_1 + t_2$ is the period of the nanostructure and n_1 and n_2 are the unknown local refractive indices. A backscattering scanning electron microscope image revealed that $T \sim 300$ nm and $f = T/t_1 < 0.2$ for Fresnel zone A. Such a grating, whose periodicity is smaller than the wavelength of the incident radiation, will produce no diffracted orders and will act as a birefringent material. When a monochromatic wave is incident upon the grating, the average index of refraction that the light will see will depend on the

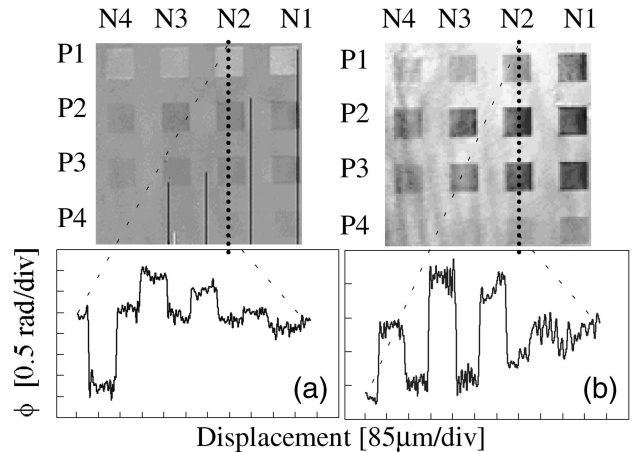


Fig. 3. Phase images of the directly written sample for two polarizations, (a) xx and (b) yy , of the light traveling into the interferometer.

Table 1. Phase changes of the Micromachined Zones

Average Power	$\Delta\phi_{xx}$ (rad)				$\Delta\phi_{xy}$ (rad)			
	N4	N3	N2	N1	N4	N3	N2	N1
P1	-0.8	-0.8	-1.6	-2.0	0.1	0.5	0.8	1.3
P2	0.8	1.2	1.2	1.2	0.7	1.2	1.4	1.5
P3	0.6	0.8	1.0	0.8	0.5	1.0	1.2	1.2
P4	0.1	0.1	0.2	0.4	0.1	0.2	0.4	0.5

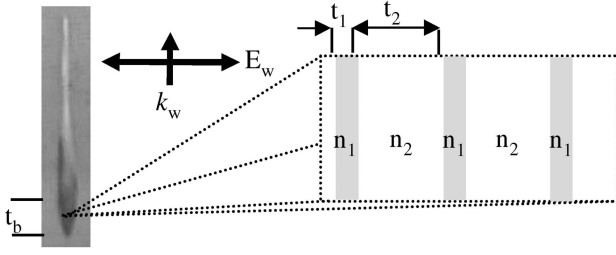


Fig. 4. Schematic of the self-organized nanograting induced in the glass. E_w and k_w , electric field and wave vector, respectively, of the laser used to micromachine the glass.

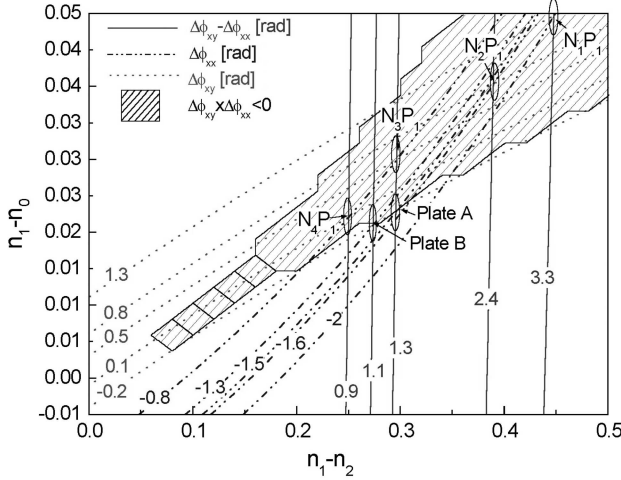


Fig. 5. Theoretical results when the model of form birefringence was applied to the directly written structures.

polarization direction of the light.¹² Figure 4 shows the typical shape of a femtosecond directly written structure in the same plane as of the propagation of the light. In agreement with what was reported by other authors,⁴ we found that only a small percentage t_b of the overall thickness contributes to the birefringence; hence the model assumes that the nanograting is localized in t_b . The phase shift measured with the interferometer is therefore given by

$$\Delta\phi_{xx} = \frac{2\pi}{\lambda} t_b \left\{ \left[\frac{n_1^2 n_2^2}{f n_2^2 + (1-f)n_1^2} \right]^{1/2} - n_0 \right\} + \overline{\Delta\phi}, \quad (2)$$

$$\Delta\phi_{xy} = \frac{2\pi}{\lambda} t_b \{ [f n_1^2 + (1-f)n_2^2]^{1/2} - n_0 \} + \overline{\Delta\phi}, \quad (3)$$

where λ is the wavelength used in the interferometer, n_0 is the index of refraction of the unprocessed material, and $\overline{\Delta\phi}$ is the phase shift given by the nonbirefringent tail of the written feature.

Figure 5 shows in a contour plot the values of $\Delta\phi_{xx}$ (dashed-dotted curve), $\Delta\phi_{xy}$ (dotted curve), and $b = \Delta\phi_{xy} - \Delta\phi_{xx}$ (solid curve) calculated for $f = 0.1$

and assuming that $t_b = 10 \mu\text{m}$ and $\overline{\Delta\phi} = 0.1 \text{ rad}$. These functions are plotted versus the local refractive-index changes. The shaded area contains all the solutions for which $\Delta\phi_{xx} < 0$ and $\Delta\phi_{xy} > 0$. The curves that correspond to various values of birefringence b are almost vertical, suggesting that the birefringence depends mainly on the difference between n_1 and n_2 . All the experimental solutions are highlighted by ellipses in the figure sized to include a variation in $\Delta\phi$ of as much as 1 order of magnitude larger or smaller. Assuming reasonable numbers, we found that the experimental results match the theory for $n_2 - n_0 = (2 \text{ to } 5) \times 10^{-2}$ and $n_1 - n_0 = (-4 \text{ to } -2) \times 10^{-1}$, and we can therefore conclude that locally the laser induces a periodic series of nanozones in which the material is denser than the substrate that alternates with other zones in which the material is more rarefied. More investigations are required, but for the first time to our knowledge a consistent model has been offered to explain birefringence, anisotropic reflection, and the negative index change shown by a directly written structure above a certain intensity threshold (type 2).

Furthermore, the results shown in Table 1 demonstrate that a difference in phase of $\sim\pi$ between the effective phase shifts seen by two orthogonal polarizations was achieved. These properties may be used to fabricate embedded half-wave plates and polarization-dependent devices such as retarders.

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